Lattice Instability in the Spin-Ladder System under Magnetic Field

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Abstract

We study theoretically the lattice instability in the spin gap systems under magnetic field. With the magnetic field larger than a critical value h_{c1}, the spin gap is collapsed and the magnetization arises. We found that the lattice distortion occurs in the spin-ladder at an incommensurate wavevector corresponding to the magnetization, while it does not occur in the Haldane system. At low temperatures the magnetization curve shows a first order phase transition with this lattice distortion.

The one-dimensional (1D) quantum spin systems with the gap in the excitation spectrum have been attracting intensive interest both theoretically and experimentally. This is due to the underlying large quantum fluctuations which destroy the magnetic long range order (LRO) [1]. Another aspect of these spin gap systems is that they are free from the instabilities, e.g., spin-Peierls instability [2], which comes from the diverging generalized susceptibility as the temperature $T \to 0$. One of the spin gap systems of the current interest is the antiferromagnetically coupled two-leg spin ladder system [3–7]. Under a magnetic field at $T = 0$ the magnetization remains zero until h reaches a critical value h_{c1} because the ground state is the spin singlet. At $h = h_{c1}$ the singlet and lowest triplet states are degenerate, and the magnetization begins to appear. Between h_{c1} and h_{c2}, where the magnetization increases, the system remains gapless and is described as a Tomonaga-Luttinger liquid with the continuously changing exponent. For $h > h_{c2}$ the system is perfectly spin polarized. Recently the two-leg ladder spin system Cu_2(1,4-Diazacycloheptane)$_2$$Cl_4$ has been studied under magnetic field [6,8–11]. The magnetization process has been studied also theoretically [12–14]. In addition, an anomaly of the specific heat is observed at $T = 1K$ when $h = 9.0T > h_{c1} = 7.2T$ [9]. This strongly suggests the phase transition to the ordered state, which is assumed to be AF long range ordering [6], but is not yet confirmed.

Motivated by these experimental works, we study in this letter the alternative instability, i.e., lattice instability, of the two-leg spin ladder system in its gapless region $h_{c1} < h < h_{c2}$. We found that when coupled to the lattice distortion, which should be always there, the gapless state is unstable to the (generalized) spin-Peierls state accompanied with the modulation of the inter-chain exchange interaction with the incommensurate wavenumber proportional to the magnetization. Another consequence of this instability is that the magnetization curve at low temperature shows a first order phase transition, and the schematic phase diagram is given in Fig. 1. This is in sharp contrast to the Haldane system, where there is no such an instability.

We start with the bosonized Hamiltonian for the two coupled spin 1/2 Heisenberg model [15–18].

$$H = H_+ + H_- + H_{\rm lattice} \tag{1}$$

$$H_+ = \int dx \Big[\frac{v_s}{2}(\Pi_+^2 + (\partial_x \phi_+)^2) - \frac{m}{\pi a_0}\cos\sqrt{4\pi}\phi_+ - \frac{h\partial_x\phi_+}{\sqrt{\pi}} \Big] \tag{2}$$

$$\begin{aligned} H_- = \int dx \Big[& \frac{v_s}{2}(\Pi_-^2 + (\partial_x \phi_-)^2) + \frac{m}{\pi a_0}\cos\sqrt{4\pi}\phi_- \\ & + \frac{2m}{\pi a_0}\cos\sqrt{4\pi}\theta_- \Big] \end{aligned} \tag{3}$$

where ϕ_j and θ_j are canonical conjugate phase fields for j-th chain, and $\phi_\pm = (\phi_1 \pm \phi_2)/\sqrt{2}$, $\theta_\pm = (\theta_1 \pm \theta_2)/\sqrt{2}$. v_s is the spin wave velocity, and a_0 is the lattice constant which we put to be 1. The mass m represents the spin gap, which originates from the inter-chain exchange interaction $J_\perp$, i.e.,

$$m \cong \frac{J_\perp}{2\pi} \tag{4}$$

Antiferromagnetic $J_\perp$ corresponds to the two-leg spin ladder, while the (strong) ferromagnetic $J_\perp$ to the Haldane system. The magnetization density $M(x)$, which is coupled to the magnetic field h, is given by the symmetric part as $M(x) = \partial_x\phi_+(x)/\sqrt{\pi}$. It is known that both H_+ and H_- have the massive spectra when $h = 0$. Only H_+ is modified by the magnetic field h and becomes gapless, while the antisymmetric part H_- remains unchanged. The coupling to the lattice distortion is given by

$$\begin{aligned} H_{\rm lattice} = & \int dx \sum_{j=1,2} \Big[\frac{1}{2}u_j(x)^2 - \frac{g}{\pi a_0}u_j(x)\sin\sqrt{2\pi}\phi_j(x) \Big] \\ & + \int dx \Big[\frac{1}{2}v(x)^2 - \frac{\gamma}{\pi a_0}v(x)[-\cos\sqrt{4\pi}\phi_+(x) \\ & + \cos\sqrt{4\pi}\phi_-(x) + 2\cos\sqrt{4\pi}\theta_-(x)] \Big] \end{aligned} \tag{5}$$

where u_j represents the dimerization of the j-th chain which modulates the intrachain exchange interaction, while v the change of the interchain distance which modulates the interchain exchange interaction. We treat the fields u_j, v classically, which is justified because the lattice is usually three-dimensional. It can be seen from eq.(2) that the magnetic field will introduce solitons when the magnetic energy gain is larger than the soliton formation energy. To make it more explicit we transform H_+ and the coupling to

the interchain modulation v into the Fermionic form by introducing the Fermion operator $\psi_{R,L}(x) = (2\pi a_0)^{-1/2}\exp[\pm i\sqrt{\pi}(\phi_+ \mp \theta_+)]$ [18]:

$$
\begin{aligned}
H_+^{\rm Fermion} = \int dx \Big[& -iv_s(\psi_R^\dagger \partial_x \psi_R - \psi_L^\dagger \partial_x \psi_L) \\
& - i(m + \gamma v(x))(\psi_R^\dagger \psi_L - \psi_L^\dagger \psi_R) + \frac{1}{2}v(x)^2 \\
& - h(\psi_R^\dagger \psi_R + \psi_L^\dagger \psi_L)\Big]
\end{aligned}
\tag{6}
$$

This transformation is possible because H_+ describes the sine-Gordon model at $\beta^2 = 4\pi$ [18]. Equation (6) represents the Dirac Fermion with the mass m coupled to the lattice distortion $v(x)$ with the Fermi energy controlled by the magentic field h. When h increases beyond $h_{c1} = m$, the Fermi level lies in the upper band of the Dirac Fermion. The magnetization M is represented by the Fermi wavenumber k_F as $k_F = \pi M$ where M is the normalized to be unity when all the spins are polarized. Then the generalized spin-Peierls transition occurs at the nesting wavevector $Q = 2k_F = 2\pi M$, i.e.,

$$
v(x) = 2v_0 \cos(2\pi M x + \delta) \tag{7}
$$

with δ being some phase. The displacement is schematically shown in Fig. 2. First consider the case where the magnetization M is not too small and the Fermi energy ε_F is large enough compared with the transition temperature T_c. A mean field treatment results in a second order phase transiton at $T = T_c \cong \varepsilon_F e^{-1/(N(\varepsilon_F)\gamma^2)}$ with the specific heat jump $\Delta C/C(T_c) = 1.42$ where $C(T)$ is proportional to T. The validity of this weak coupling mean field theory can be examined as follows. The experiment has been done for $h = 9.0T$ where the magnetization $M \cong 0.3$ [6,9,10]. The gap $m \cong 0.89meV$ and the spin wave velocity v_s is estimated from the intrachain exchange interaction $J_2 = 0.21meV$ as $v_s = \frac{\pi}{2}J_2 \cong 0.33meV$ [9]. Therefore $v_s k_F = \pi v_s M \cong 0.3meV$, which is about one-third of m. The Fermi energy ε_F measured from the bottom of the upper band is then $\varepsilon_F \cong 0.05meV \cong 0.5K$. This estimate shows that the transition temperature T_c is the same order as the Fermi energy ε_F, and the system is in the intermediate coupling regime.

As the magnetic field is decreased to h_{c1} the Fermi energy ε_F also decreases and the mean

field treatment breaks down. Then we consider the zero temperature limit with changing the magnetic field. When the magnetization M is small and the off-diagonal matrix element γv_0 introduced by the distortion is larger than the Fermi energy ε_F, many k points are connected by that matrix element. This is due to the parabolic dispersion which becomes important near the bottom of the band. As shown below the kinetic energy gain due to the distortion is given by $4\pi M\gamma v_0$ and v_0 is determined as $v_0 = 2\pi\gamma M$. With this v_0, the matrix element is $2\pi\gamma^2 M$ and equating this to the dispersion $(v_s k_0)^2/2m$ we obtain $k_0 \cong \sqrt{4\pi m\gamma^2 M}/v_s$ The number G of the reduced Brillouin zones within the region $[-k_0, k_0]$ is $G \cong k_0/(\pi M) \cong \sqrt{4m\gamma^2/(\pi v_s^2 M)}$ and $G >> 1$ in the limit of small M. In this case we can neglect the dispersion $(v_s k)^2/2m$ in the diagonal matrix element compared with the off-diagonal ones, and the eigen-value problem can be approximately regarded as that for the tight binding wave with the wavenumber q discretized as $q_n = 2\pi n/G$ (n: integer). Then the eigen-value is given by

$$\varepsilon_n = -2v_0\gamma\cos q_n. \tag{8}$$

Therefore the lowest band ($q = n = 0$) with the energy $-2\gamma v_0$ is occupied and the energy gain is $-4\pi\gamma v_0 M$. Then minimizing $v_0^2 - 4\pi\gamma v_0 M$, we obtain $v_0 = 2\pi\gamma M$ as mentioned above. The gap between the lowest ($n = 0$) and the second lowest one ($n = 1$) is of the order of $v_0\gamma/G^2 \propto M^2$ and is much smaller than the matrix element $v_0\gamma(\propto M)$ in the limit of small M. This happens when the magnetic field h is slightly above the critical field $h_{c1}^{(0)}$ without the lattice distortion and we define $\Delta h = h - h_{c1}^{(0)}$. Based on the above discussion, let us consider Δh as a function of the magnetization M. Without the lattice distortion, $\Delta h = (\pi v_s M)^2/(2m)$ for small M. With the coupling to the lattice, it changes into

$$\Delta h = -2\gamma v_0 + \alpha(\pi v_s M)^2/m = -4\pi\gamma^2 M + \alpha(\pi v_s M)^2/m \tag{9}$$

where α is a constant of the order of unity. This is because the chemical potential at $T = 0K$ is at the middle of the gap between the lowest ($n = 0$) and the second lowest ($n = 1$) bands which is $-2\gamma v_0 + O((v_s M)^2/m)$. The relation (9) means that the magnetization M jumps

from 0 to a finite value $M = M_0 \sim m\gamma^2/v_s^2$ with a first order phase transition. Summarizing the considerations above, the schematic phase diagram for spin ladder system coupled to the lattice is given in Fig. 1. The first order phase transition line terminates at a tricritical point to change into the second order transition line.

There is no instability for the dimerization u_j, which corresponds to the standard spin-Peierls distortion, when the coupling constant g is small enough. This is because the operator $\sin\sqrt{2\pi}\phi_j = \sin\sqrt{\pi}(\phi_+ \pm \phi_-)$ contains $\cos\sqrt{\pi}\phi_-$ and $\sin\sqrt{\pi}\phi_-$. In ref. [18] it is discussed via the mapping the Ising model that the correlation functions for $\cos\sqrt{\pi}\phi_-$ and $\sin\sqrt{\pi}\phi_-$ decay exponentially. Then also the correlation function of $\sin\sqrt{2\pi}\phi_j$ decays exponentially. This means that the energy gain due to the dimerization u_j is given by $\sim g^2u_j^2$ for small g even at zero temperature, and the system is stable against the dimerization as long as g is small because of the elastic energy $u_j^2/2$. Therefore, the lattice instabillity does not occur in the Haldane system where the interchain coupling $J_\perp$ represents the ferromagnetic Hund's coupling and is very strong ($\sim 1eV$). The spin gap is due to the quantum fluctuation of the resulting $S = 1$ spins [1], and the modulation of $J_\perp$ does not play any role. Then the magnetization process can be discussed by a model without the coupling to the lattice [19,20]. Another instability is of course the AF ordering as discussed in ref. [6]. The competition between the lattice instability and the AF is determined by the relative magnitude of the inter-ladder exchange interactions and the spin-lattice coupling, and the coexistence of these two is not expected for clean system [21].

We now discuss the experimental consequences of the above scenario. (i) The Bragg spot should be observed in the X-ray and/or neutron scattering at the wavevector $Q = 2\pi M$ due to the modulation of the interchain distance. (ii) There appears a gap below T_c and the specific heat should behave as $C \sim e^{-\gamma v_0/T}$, where γv_0 is the gap of the order of the transition temperature T_c. (iii) The magnetization curve shows a hysteresis behavior at low temperatures. The experimental confirmation of these predictions is highly desired.

In conclusion we have studied the lattice instability in the spin-ladder system when the spin gap is collapsed by the magnetic field. The lattice will distort to modulate the interchain

exchange interaction with the incommensurate wavenumber $Q = 2\pi M$.

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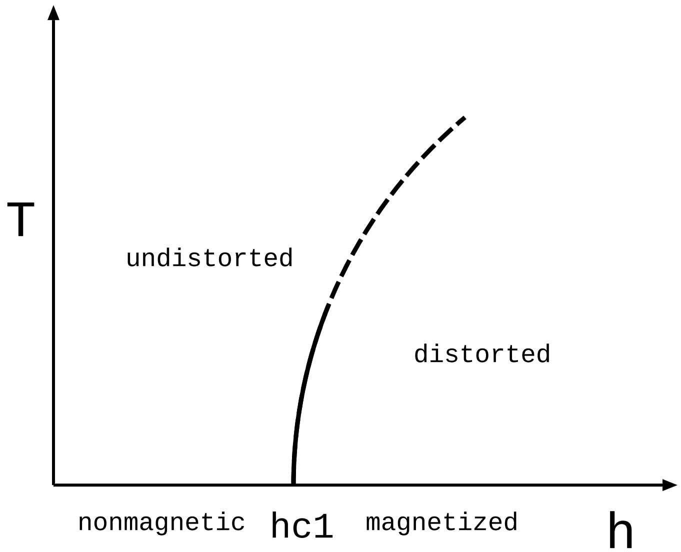
Figure captions

Fig. 1: Schematic phase diagram of the two-leg spin ladder system coupled to the lattice distortion in the plane of temperature T and the magnetic field h. The thick solid curve is the first order phase transition line while the broken one the second order transition between the undistorted and distorted states. The magnetization jumps across the first order transition line.

Fig. 2: Schematic view of the lattice distortion which modulates the interchain exchange interaction. The wavenumber $Q = 2\pi M$ changes as the magnetization M increases.


T
undistorted
distorted
nonmagnetic
hc1
magnetized
h